

REMARKS

The rejection of Claims 1, 2 and 4-13 under 35 U.S.C. §103(a) as unpatentable over EP 792858 (Takagawa et al) in view of either U.S. 3,957,896 (Yokohama et al) or U.S. 6,057,487 (Munson et al), is respectively traversed.

The present invention relates to a process which, on the basis of suitable crystallization and filtration techniques, allows the separation of 2,6-dimethylnaphthalene (2,6-DMN) from mixtures in which it is present with isomers thereof.

As described in the specification beginning at page 1, line 15, 2,6-DMN has been synthesized by many different processes in the prior art, often resulting as part of a mixture containing isomers thereof and other compounds. Various techniques have been used to separate 2,6-DMN from such mixtures. Such techniques have been problematical. Two phenomena complicate the separation of high purity 2,6-DMN: one, the crystal obtained by crystallization from the molten state has such a morphology that, after separation of the mother liquor by filtration, the residual wetting of the solid is high and therefore the titer of 2,6-DMN in the solid is low; and two, the presence of a co-crystallization phenomenon due to the presence of other compounds in the solid, and particularly, 2,7-DMN. For example, the 2,7/2,6 ratio in the solid is about 10% of this ratio in the mother liquor. Solutions to this problem have been suggested in the prior art, but these are also problematical.

The present invention addresses the problems of the prior art and is surprisingly simple and effective, and results in the production of 2,6-DMN having a very high degree of purity, with contemporary recoveries close to the theoretical value, without any of the limits and disadvantages which characterize the methods described in the state-of-the-art.

As recited in Claim 1, the present invention is a process for the separation of 2,6-dimethylnaphthalene from a starting mixture containing it and isomers thereof comprising the following operations: (A) crystallization of 2,6-dimethylnaphthalene by the addition of a

solvent and cooling of the mixture thus obtained to a temperature higher than the highest formation value of any eutectic of 2,6-dimethylnaphthalene and another isomer in the mixture, whereby a mother liquor containing solid, said solid comprising 2,6-dimethylnaphthalene, is produced; (B) removal of the mother liquor by repeated washings with a solvent; (C) dissolution in a solvent of the solid obtained, whereby a solution is produced; (D) crystallization of said solution by cooling, whereby a suspension is produced; (E) filtration of said suspension, whereby said 2,6-dimethylnaphthalene is separated, and wherein the solvent used for each of operations (A), (B) and (C), is independently selected from the group consisting of low molecular weight aliphatic alcohols, glycols, and mixtures thereof.

In a preferred embodiment, the same solvent is used for each of operations (A), (B) and (C), and more preferably, is methanol. When such a solvent is used, the above-discussed co-crystallization is substantially reduced in degree and consequently, the isomers present as solid in the 2,6-DMN crystal are in a lower amount. For example, the 2,7/2,6 ratio in the solid is about 2% of the 2,7/2,6 ratio in the mother liquor.

In sum, the present invention is a process to obtain pure 2,6-DMN from mixtures containing it with other isomers, comprising the following fundamental steps:

- (A) crystallization by addition of a low molecular weight aliphatic alcohol or glycol solvent (alcoholic solvent), preferably methanol, and cooling of the mixture up to a temperature higher than that of formation of the eutectic;
- (B) removal from the crystals of the residual mother liquors by washing with a solvent of the same type as, and preferably the same as, step (A);
- (C) dissolution of the crystals coming from step (B) in a solvent of the same type as, and preferably the same as, step (A);
- (D-E) crystallization by cooling and filtering.

Step (A)

In step (A), the use of the alcoholic solvent surprisingly results in reducing the phenomenon of co-crystallization. On the contrary, crystallization in the absence of solvent, including cooling to a temperature higher than that of precipitation of the eutectic, results in significantly more co-crystallization, namely the inglobation of other isomers, in particular of 2,7-DMN into the crystals of 2,6-DMN.

The result is that crystallization carried out in the absence of solvent gives a solid having a 2,7/2,6 ratio corresponding to 10% of the value of the same 2,7/2,6 ratio measured in the corresponding crystallization mother liquors; a crystallization of the same isomers mixtures using methanol as crystallization solvent gives a solid having a 2,7/2,6 ratio corresponding to 2% of the value of the same 2,7/2,6 ratio measured in the corresponding crystallization mother liquors, as described above.

Step (B)

In step (B), a relevant amount of mother liquor residual is removed by washing with alcoholic solvent capable of removing undesired isomers contained in the mother liquor without dissolving the solid.

Steps (C)-(E)

In steps (C)-(E), 2,6 DMN crystals are dissolved again in an alcoholic solvent and recrystallized. The alcoholic solvent controls co-crystallization, allowing the production of an improved purity of 2,6 DMN.

Takagawa et al, which was cited in the corresponding European Search Report as “A”, i.e., technological background, category only, discloses a process for the production of highly pure 2,6-DMN by crystallization from a mixture of DMN isomers. Takagawa et al disclose aliphatic saturated hydrocarbons and alicyclic saturated hydrocarbons only as the solvent for use in their process (page 5, lines 19-20). Takagawa et al require various compositional limitations in their starting mixture (page 6, line 19 ff), particularly that it contain at most 10% by weight of 2,7-DMN (page 7, lines 10-12). Indeed, the examples in Takagawa et al confirm that their process is applicable only with relatively small amounts of 2,7-DMN. Either the amount of 2,7 isomer is so small as to not even be quantified or, as in Examples 5 and 6, it is less than 5 wt%. The present invention, on the other hand, is not so limited and indeed, the Example herein, described beginning in the specification at page 10, line 10, uses a starting mixture having a concentration of 2,7-DMN of 15.9 wt%.

While Applicants appreciate that the presently-claimed process is not limited to one requiring a minimum amount of 2,7-DMN greater than the 10 wt% 2,7-DMN maximum of Takagawa et al, nevertheless, this difference highlights the fact that besides the difference in solvents, the presently-claimed invention is otherwise different from the crystallization process of Takagawa et al. Note that the only limitation herein with regard to a starting mixture containing 2,7-DMN isomer is that recited in Claim 2, i.e., the 2,6 isomer be present in an amount higher than that of its eutectic with the other isomers.

Takagawa et al further disclose that the crystallizing operation temperature is preferably 10-60°C, and is correlated with the amount of solvent to be used, the solubility of 2,6-DMN in the solvent, the concentration of 2,6-DMN in the mixture of DMN isomers, and the concentration of the suspension at the end of the crystallization (page 7, lines 23-25).

Notwithstanding all that has been discussed above, Takagawa et al neither disclose nor suggest the presently-recited combination of steps (A)-(E) of Claim 1 herein, notwithstanding differences in solvent.

At page 3, lines 13-15 of the Office Action, the Examiner finds that "The crystal is then dissolved in solvent to produce a solution which is passed into a recrystallization unit to produce a suspension which is filtered to obtain a pure 2,6-DMN product."

Contrary to the Examiner's opinion, it is submitted that Takagawa et al does not disclose or suggest a step of further dissolution of the crystals resulting from the crystallization step, in a solvent such as the crystallization solvent, followed by a crystallization by cooling and filtering. Rather, Takagawa et al is drawn only to a method to accelerate the crystals growing up, as disclosed at page 10, lines 32-35. The method, once the precipitation of 2,6-DMN crystals is obtained, involves causing further dissolution of a part of the crystals by mild heating, crystals that then re-precipitate by further cooling. The explanation of this acceleration effect is provided at page 10, lines 44-48, and an application of this procedure is exemplified in Example 14, lines 46-47 and in Example 20, lines 38-40.

This kind of "partial" recrystallization having the goal of accelerating the growth of the crystals is not suggestive in any way the step of recrystallization according to the present invention, having the goal of obtaining pure 2,6-DMN crystals.

Recognizing the deficiencies in Takagawa et al, the Examiner relies on Yokoyama et al and Munson et al. Neither reference remedies these deficiencies.

Yokoyama et al discloses a process for preparing 2,6-dimethylnaphthalene from a mixture of DMN isomers which involves, *inter alia*, adjusting the weight ratio to not more than 12 of the combination of 2,6-DMN, 1,6-DMN, and 1,5-DMN (group A), to other DMNs, and the weight ratio of 2,6-DMN to at least 27 of group A. Particularly, the process involves selective isomerization of a DMN mixture, crystallization to separate 2,6-DMN, distillation to

separate the low-boiling or the high-boiling components of the DMNs at any stage during the process, and recycle of the crystallization residue to the isomerization step.

The crystallization step is not particularly limited. For example, 2,6-DMN can be obtained through crystallization at a temperature higher than that of formation of the eutectic mixture, without addition of further solvents to the reaction product in the isomerization step. If the obtained purity is lower than desired, crystallizations and washings are repeated to reach the desired purity. Solvents such as alcohols, aromatic hydrocarbons and aliphatic hydrocarbons are disclosed as useful in the crystallization step.

The crystallization is preferably carried out without adding further solvents at a temperature higher than that at which the eutectic mixture forms. Indeed, all the examples are carried out in the absence of solvent. Thus, not only does Yokoyama et al not specify any preference among the various solvents disclosed, Yokoyama et al prefers **no** solvent be used.

The Examiner relies on Yokoyama et al for their disclosure of crystallization carried out at a temperature higher than that at which a eutectic mixture forms. However, to that extent, Yokoyama et al adds nothing to what Applicants have already acknowledged is known, as disclosed in the specification at page 4, lines 2-9. Everything else in Yokoyama et al teaches against the present invention.

Munson et al is drawn to a method for producing 2,6-DMN from mixed dimethylnaphthalenes by crystallization, adsorption and isomerization. Particularly, Munson et al disclose a method of purifying 2,6-DMN from a feed mixture of DMN isomers and near-boiling compounds comprising the steps of crystallizing the mixture to precipitate a eutectic composition comprising 2,6-DMN and 2,7-DMN; optionally dissolving the eutectic composition in a solvent; and recovering a predominantly 2,6-DMN composition from the dissolved eutectic composition by adsorbing out non-2,6-DMNs onto an adsorption column

(column 3, lines 20-30). As a means for separating the 2,6-DMN/2,7-DMN eutectic, Munson et al disclose solvent crystallization using a solvent which includes, *inter alia*, the hydrocarbon solvents of Takagawa et al and the presently-recited alcohols (column 5, line 49 ff).

The Examiner finds that in view of the above-discussed disclosure with regard to solvents, it would have been obvious to one of ordinary skill in the art to substitute the hydrocarbon solvent of Takagawa et al with the alcohol solvent of Munson et al or Yokoyama et al. However, this finding ignores essential differences between Takagawa et al, on the one hand, and Yokoyama et al and Munson et al, on the other hand, such that one skilled in the art would not have made this substitution, for reasons discussed below. However, prior to discussing these differences, it is important to keep in mind the basic discovery herein, as now reiterated, which is the finding of a sequence of steps that, all together, cause the production of 2,6-DMN from mixtures containing it, with high purity and yield, and in the finding of a class of alcohols, particularly methanol, and of glycols as solvents for the crystallization of 2,6-DMN from mixtures of its isomers.

In Takagawa et al, the initial crystallization of 2,6-DMN is with the use of a solvent. In Munson et al, it is disclosed that crystallization of the eutectic be carried out in the absence of a solvent when the concentration of 2,6-DMN and 2,7-DMN isomers is higher than 20% and more preferably higher than 90% (column 5, lines 29-32), while a solvent should be used only when the concentration of 2,6-DMN and 2,7-DMN isomers is low (column 5, lines 49-52). Thus, Munson et al suggests that a DMN starting mixture of the type disclosed in Takagawa et al (when the concentration of 2,6-DMN and 2,7-DMN isomers is higher than 20%) should be subjected to a melt crystallization, i.e., a crystallization without a solvent. In other words, the disclosure of solvents in Munson et al would not be considered to be relevant to the DMN compositional starting mixtures of Takagawa et al.

Moreover, even if Takagawa et al and Munson et al were combined, the result would not be the presently-claimed invention, because the present invention does not seek to precipitate a eutectic composition comprising 2,6-DMN and 2,7-DMN. Indeed, step (A) of Claim 1 crystallizes down to a temperature which is **higher than** the formation value of such a eutectic.

In addition, while Munson et al is concerned with starting mixtures having a significant amount of 2,7-DMN with regard to 2,6-DMN, Takagawa et al relates more to starting mixtures containing very little relative amount of 2,7-DMN, as discussed above.

Further, Munson et al requires the use of an adsorbent to separate 2,6-DMN from 2,7-DMN. On the contrary, the present invention does not involve the use of adsorbents.

Finally, it is noted that Example 4 of Munson et al, which uses meta-xylene as a crystallization solvent, produces a crystallization yield of 16.4%, which is significantly lower than that obtainable by the presently-claimed invention.

In sum, Munson et al addresses the problem of recovering 2,6-DMN having a high purity degree from mixtures containing it with other isomers, through the use of an adsorbent. Before the passage through the adsorbent, the mixture is submitted to a crystallization treatment allowing the recovering of said mixture from the eutectic formed by 2,6 and 2,7 DMN. In Munson et al, the crystallization of their first step produces a precipitate containing both the 2,6 and 2,7 isomers of DMN, while other impurities remain in the supernatant (column 3, lines 19-24; column 4, lines 57-59).

While Munson et al discloses a wide variety of solvents--hydrocarbons such as toluene, xylene, octane and heptane, alcohols such as methanol, ethanol, and isopropanol, ethers, carboxylic acids and acetic acid, and combinations thereof--to separate the 2,6-DMN and 2,7-DMN eutectic from the mixture containing it, no such solvents are disclosed for separation of the 2,6-DMN isomer **from** the 2,7-DMN isomer.



Indeed, it would be contraindicated to use a solvent able to precipitate the 2,6 and 2,7-DMN eutectic, to obtain precipitation of pure 2,6-DMN only from a mixture containing it together with 2,7-DMN.

Examples 2 and 4 of Munson et al highlight their "coarse crystallization step to reduce the level of non 2,6/2,7 isomers" (column 8, lines 53-55), which is "followed by adsorption where the feed has a high proportion of 2,6-DMN" (column 9, lines 55-57).

Thus, Takagawa et al and Munson et al go in opposite directions; Munson et al discloses precipitating by crystallization both 2,6 and 2,7-DMN isomers from a mixture containing them together with other isomers and impurities, while Takagawa et al is drawn to purified 2,6-DMN alone while 2,7-DMN remains in the crystallization mother liquor. Indeed, Munson et al teach against the presently-claimed condition that crystallization be carried out to a temperature higher than the precipitation temperature of the first eutectic of 2,6-DMN.

Thus, Applicants respectfully submit that it is not *prima facie* obvious to substitute any of the presently-recited solvents for the solvents of Takagawa et al. Nevertheless, the newly-submitted Bignazzi Declaration provides declaratory support for the proposition that the present solvents are superior to those of Takagawa et al. A mathematical model was used to compare the results obtained in Example 1 of Takagawa et al and results obtainable using the same crystallization temperature and the same isomer mixtures, but with methanol used as the solvent. The mathematical model simulated the crystallization at 20°C of the Takagawa et al's Example 1, carried out in heptane, and in methanol, as shown in Tables A and B, respectively, submitted with the amendment filed October 24, 2002, and attached in the Bignazzi Declaration. By comparing the results in Tables A and B, it can be seen that the crystallization yield of 2,6 DMN significantly increases when changing from heptane

(66.4%) to methanol (80.9%). In addition, the purity in the panel remains practically unchanged, i.e., 99.31% using heptane, and 99.34% using methanol.

In the Office Action, the Examiner finds (page 4, last paragraph) that the claimed process does not include the limitation of the amount of 2,7-DMN in the feed mixture.

In reply, it has been demonstrated that an alcoholic solvent, such as methanol, behaves differently, when used in the crystallization, compared to other solvents, because it achieves recovery of 2,6-DMN in the presence of **any** amount of 2,7-DMN, at the condition that the 2,6-DMN amount is greater than the eutectic. It is also evident that the process of the present invention provides improved results that could not have been predicted by the applied prior art. In fact, all comparisons between the present invention and the examples of Takagawa et al, at the same conditions with high and low amounts of 2,7-DMN, indicate higher yields to 2,6-DMN having the same or improved purity.

The Examiner also finds (page 5, first paragraph) that the temperature range of the present invention does not distinguish over the range in Takagawa et al. In reply, it is emphasized that the presently-claimed process is not different from the process of Takagawa et al because of the crystallization temperature but because of the results that are obtained at such temperature. In fact, using an alcoholic solvent such as methanol, high yields are obtained and co-crystallization is very limited.

The Examiner equates alcohols such as methanol with aliphatic hydrocarbons in the 2,6-DMN crystallization process. According to the present invention, it has been very surprisingly found that an alcoholic solvent such as methanol has a definitely different behavior. In fact, as already indicated, it gives a higher yield to 2,6-DMN and a higher purity in crystallization. Also in the washing steps an alcoholic solvent such as methanol presents some advantages; in fact, methanol, although being completely miscible with mother liquors at the working temperature range, very poorly dissolves the 2,6-DMN formed crystals. The

advantageous action of methanol with respect to the other solvents is evident also in the recrystallization. In fact, it surprisingly allows to obtain high yields and purities.

Regarding the Examiner's finding (page 5, second paragraph) in response to the argument that Munson et al uses solvent when the concentration of 2,6-DMN and 2,7-DMN is low whereas the concentration of 2,6-DMN and 2,7-DMN in Takagawa et al is high, it has been responded to above.

Regarding the Examiner's findings (page 5, third paragraph and paragraph bridging pages 5 and 6) in response to various arguments, the Examiner explains how he applied the prior art. In so doing, the Examiner makes clear that he did not properly apply section 103 of the statute, which requires that the subject matter **as a whole** be considered. The Examiner has extracted from the prior art only that which he believes supports his position while ignoring other contrary teachings. First of all, none of the applied prior art supports the Examiner's position, for reasons detailed above. Second of all, the parts of the applied prior art ignored actually teach away from the present invention.

For all the above reasons, it is respectfully requested that the rejection be withdrawn.

**Applicants respectfully traverse the finality of the Office Action.** In the amendment filed October 24, 2002, Claim 1 was amended by incorporating the subject matter of Claim 3 therein. All other additional amendments to Claim 1 were made to overcome formal rejections, but no further change in claim scope was effected. Claim 3 was previously rejected over the combination of Takagawa et al and Munson et al only. Thus, the reliance now on Yokoyama et al in the first instance to reject present Claim 1 was **not** necessitated by Applicants' amendments. Indeed, as provided by M.P.E.P. § 706.07(a), a second or any subsequent action on the merits will not be made final if it includes a rejection on newly cited art of any claim not amended by Applicants in spite of the fact that other claims may have been amended to require newly cited art. In effect, present Claim 1 is of the same scope as

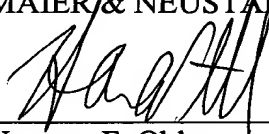
Application No. 09/851,131  
Reply to Office Action of

original Claim 3. Accordingly, it is respectfully requested that if the present response does not result in a Notice of Allowance, the finality of the Office Action be withdrawn.

All of the presently pending claims in this application are now believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,  
MAIER & NEUSTADT, P.C.



---

Norman F. Oblon  
Attorney of Record  
Registration No. 24,618

Customer Number

**22850**

Tel: (703) 413-3000  
Fax: (703) 413 -2220  
(OSMMN 08/03)  
NFO/HAP/cja

Harris A. Pitlick  
Registration No. 38,779